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Influence of quasi-layer-by-layer roughness on proximity effects in thin film superconducting/normal-metal junctions

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Abstract

We investigate the influence of quasi-layer-by-layer interface roughness on proximity effects, at the junction of a thin surperconducting/normal-metal film. The reduction of the superconducting film critical temperature $\Delta T_c/T_c$ is shown to decrease with film thickness in an oscillatory manner, which reflects the presence of growth front pinning effects during roughness formation. The oscillations are significant for weak surface relaxation where $\Delta T_c/T_c$ varies significantly with film thickness, and decrease in magnitude with increasing film thickness. In contrast, in the presence of growth front instability, the pinning induced oscillations are shown to be further amplified with increasing instability strength.

Keywords: Proximity effect; Thin films; Fluctuation effects

1. Introduction

Proximity effects constitute currently a topic of intense research especially in the field of thin film superconductivity (i.e., anisotropic high-temperature Cu-oxide superconductors) [1–5]. At the junction of a normal metal and a superconducting thin film penetration of Cooper pairs from the superconductor into the metal and electrons from the normal metal into superconductor occurs, which determines the so-called proximity effects [1–3]. The latter manifest themselves by reducing the critical temperature T_c of a thin superconducting film covered by a thick normal metal film [1–3]. Indeed, for an isotropic superconducting thin film of thickness d with a flat superconductor/normal-metal (SN) the reduction of the critical temperature [6] is given by $\Delta T_c/T_c = -\gamma^2\pi^2\xi_0^2/4d^2$ with $\gamma \approx 0.74$ a numerical factor, and ($\ll$d) the zero temperature coherence length. For an anisotropic superconductor, $\Delta T_c/T_c$ depends on the relative orientation of the SN-interface relative to the superconductor symmetry axes, and becomes maximum if this interface is perpendicular to the axes corresponding to the maximum coherence length [6].

For a rough SN-interface, the relative orientation of the SN-interface with respect to the symmetry axes will vary laterally, enhancing the reduction of the critical temperature $\Delta T_c/T_c$ [6]. For a self-affine rough interface, small interface

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roughness exponents (which quantify fine roughness details at short wavelengths) H (<0.5) can strongly influence $\Delta T_c/T_c$ besides the long wavelength roughness ratio Δ/ξ with Δ the rms roughness amplitude and ξ the lateral correlation length [7]. A rough self-affine growth front arises due to competition between noise induced roughening and surface relaxation of incoming adatoms (stable growth) [8–10]. Nevertheless, under unstable growth conditions the formation of a characteristic multilayer step structure in the form of mounds takes place [11–15]. In this case, $\Delta T_c/T_c$ shows a complex dependence on the system correlation length and the average mound separation depending on their relative magnitude with respect to each other [16].

At any rate a layer-by-layer (LBL) growth would eliminate additional enhancement of proximity effects due to interface roughness. However, such a growth mode does not always occur (i.e., for small surface mobility of deposited adatoms with respect to the deposition rate). As a result some nucleation on top of already existing islands can commence which is only a small portion compared to the bottom-grown layer. Such a growth mode is termed as quasi-layer-by-layer (Q-LBL) where lattice pinning effects are significant in promoting partial LBL growth [17,18]. So far, any quantitative investigation of this type of morphology on proximity effects is still missing. This will be the topic of the present paper where calculation of roughness effects on $\Delta T_c/T_c$ will be performed for rough growth fronts associated with a non-equilibrium sine-Gordon model, which describes Q-LBL growth by incorporation of surface relaxation, noise induced roughening, and lattice pinning effects.

2. Proximity effect theory and roughness model for non-flat SN-interfaces

2.1. Proximity theory

We defined $h(r)$ ($\ll$d) as the SN-interface height fluctuation resulting from film growth with $r=(x,y)$ the in-plane position vector, and $\partial h/\partial x_i$ the partial height derivatives. The zero temperature coherence lengths ($\ll$d) along the a–b, and c-axes respectively are considered to be parallel to x–y, and z-axis ($i=x$, y, $c\equiv z$). The formula for $\Delta T_c/T_c$ by Mints and Snapiro [6] upon Fourier transformation of $h(r)$ yields for translation invariant surfaces (or $\langle h(q)h(q')\rangle = [(2\pi)^4/S]\delta^2(q-q')\langle|h(q)|^2\rangle$) [7,16]

$$\Delta T_c/T_c = -\frac{\gamma^2\pi^2\xi_c^2}{4d^2}\left\{1+\frac{(6+\pi^2)}{3\pi^2\xi_c^2}\times\left(\sum_{i=1}^{2}\xi_i^2\left[\frac{(2\pi)^4}{S}\right]\int_0^{Q_c} q_i^2\langle|h(q)|^2\rangle \mathrm{d}^2q\right)\right\} \tag{1}$$

with S the average flat interface area. Clearly in Eq. (1) the knowledge of the interface roughness spectrum $\langle|h(q)|^2\rangle$ is required to further calculate $\Delta T_c/T_c$.

2.2. Roughness model

A Langevin growth equation describing 2D-nucleation and roughening during Q-LBL growth reads of the form [17,18]

$$\frac{\partial h(r,t)}{\partial t} = (-1)^{n+1}v_n\nabla^{2n}h - \varepsilon A\sin[2\pi h(r,t)/c)] + R + n(r,t). \tag{2}$$

Eq. (2) incorporates surface relaxation by the term $(-1)^{n+1}v_n\nabla^{2n}h$ (evaporation/recondensation for $n=1$, and surface diffusion for $n=2$). $n(r,t)$ represents Gaussian random noise fluctuations, during evaporation at a rate R, of amplitude D such that $\langle n(r,t)n(r',t')\rangle = 2D\,\delta^2(r-r')\,\delta(t-t')\times(\langle n(r,t)\rangle = 0)$. A is the strength of the pinning term $V_{\text{pin}} = -\varepsilon A\,\sin[2\pi h(r,t)/c]$ that favors energetically integer values of the surface height in units of the atomic spacing c. For a small pinning term, a perturbative solution of Eq. (2) is possible (see Appendix A) [18]. After some calculation, we obtain the roughness spectrum $\langle|h(q)|^2\rangle$

$$\langle |h(q)|^2 \rangle = \frac{S}{(2\pi)^5} \left\{ D\frac{1-\mathrm{e}^{-2F(q)t}}{F(q)} + \frac{ADc}{\pi R}\frac{\mathrm{e}^{-2F(q)t}}{F(q)} \sin(t/\tau_c) - 4AD\frac{[\cos(t/\tau_c) - \mathrm{e}^{-2F(q)t}] - [\sin(t/\tau_c)/cF(q)]}{(1/\tau_c)^2 + 4F(q)^2} \right\} \quad (3)$$

with $F(q) = v_n q^{2n}$, $\tau_c = c/2\pi R$, and the film thickness $d = Rt + (A\tau_c)[\cos(t/\tau_c) - 1]$.

3. Results and discussion

Interface roughness can induce a considerable reduction of the critical transition temperature as long as the in-plane coherence lengths are significantly larger than the one out-of-plane ($\xi_i \gg \xi_c$). Indeed, under these conditions, measurements of the critical temperature T_c in slowly grown $YBa_2Cu_3O_7$–$YBa_2Cu_{3-x}Co_xO_{7+\delta}$ bilayers [5] revealed a good agreement with the former roughness theory predictions in terms of Eq. (1) [6]. Our calculations of $\Delta T_c/T_c$ were performed for evaporation rate $R = 0.3$ nm s^{-1} and noise amplitude $D = 0.01$ nm s^{-1} in order that $D \ll R$. The units of the relaxation coefficients v_n are assumed to be respectively $[v_1] = 10^{-2}$ nm^{-2} s^{-1} and $[v_2] = 10^{-4}$ nm^{-4} s^{-1}. Finally, the coherence lengths considered here during the calculations were $\xi_{x,y} = 1$ nm and $\xi_z = 0.3$ nm, as well as the film thickness d was limited to values $d \geqslant \xi_{x,y,z}$ for Eq. (1) to be valid.

Fig. 1 shows the relative critical temperature reduction $\Delta T_c/T_c$ vs. film thickness d for $n = 1$ (surface relaxation by evaporation/recondensation), and various pinning amplitudes A. For $A > 0$, the pinning effects manifests themselves as oscillations on an otherwise decreasing $\Delta T_c/T_c$ with increasing film thickness d. Such a decrease results from the fact that as the film thickness d increases, $\Delta T_c/T_c$ decreases primarily as $\Delta T_c/T_c \propto d^{-2}$ while the interface slopes $\langle(\partial h/\partial x_i)^2\rangle$ increase slower than d^2 for stable growth conditions ($v_n > 0$) [16], following an oscillatory behavior that leads finally to the observed oscillations for $\Delta T_c/T_c$.

Moreover, as Fig. 2 indicates, with increasing surface relaxation or equivalently increasing coefficient v_n, the pinning induced oscillations diminish and finally disappear for significantly large film thickness. This is because the smoothing effect from evaporation/recondensation (similarly for surface diffusion) is sufficient to suppress any roughness related to nucleation of incoming adatoms

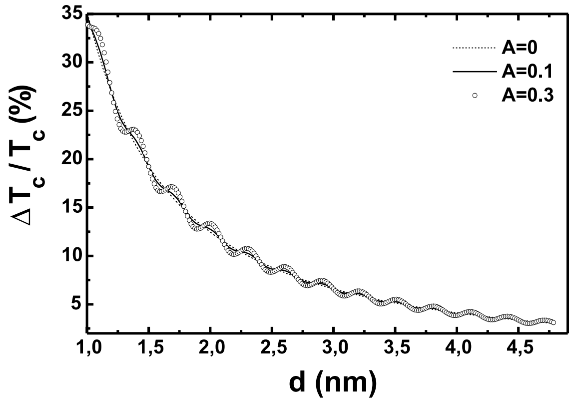

Fig. 1. $|\Delta T_c|/T_c$ vs. film thickness d for stable Q-LBL growth with surface relaxation due to evaporation/recondensation for various pinning amplitudes A as indicated and surface relaxation coefficient $v_1 = 0.1$.

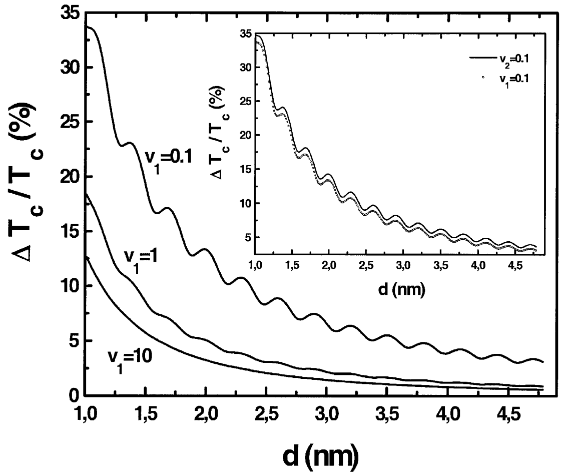

Fig. 2. $|\Delta T_c|/T_c$ vs. film thickness d for stable Q-LBL with surface relaxation due to evaporation/recondensation for pinning amplitude $A = 0.3$ and various relaxation coefficients v_1 as indicated. The inset shows $|\Delta T_c|/T_c$ vs. film thickness d for stable Q-LBL growth where comparison of surface relaxation due to evaporation/recondensation and surface diffusion is shown for $A = 0.3$ and $v_1 = v_2 = 0.1$.

on top of existing islands prior to completion of the first layer. In addition, such a smoothing effect results in lower decrement in magnitude of $\Delta T_c/T_c$, and thus of the critical temperature T_c with increasing film thickness. In addition, the inset of Fig. 2 compares the effect of the two relaxation mechanisms (surface diffusion and evaporation/recondensation) for significant pinning amplitudes. For surface diffusion which acts as an effective relaxation at smaller lateral length scales than that of evaporation/recondensation ($r < 2\pi(v_1/v_2)^{1/2}$), the pinning induced oscillations sustain at slightly larger film thickness, as well as $\Delta T_c/T_c$ appears also slightly larger in absolute magnitude.

Finally, we shall compare pinning effects on $\Delta T_c/T_c$ for the case of unstable growth where an instability is created by significant Schwoebel barriers at step edges that prevent down hill diffusion of incoming adatoms (favoring the formation of multilayer step structures in the form of mounds), with the case of stable growth with evaporation/recondensation as the main relaxation mechanism. The roughness spectrum for unstable growth is obtained from that of stable growth (with evaporation/recondensation) upon substitution $v_1 \rightarrow -v_1$ (at early stages of growth) [15].

As Fig. 3 indicates, for unstable growth the pinning induced oscillations on $\Delta T_c/T_c$ remain significant in magnitude with increasing film thickness d, while for stable growth they reduce in magnitude with increasing film thickness. For unstable growth such a behavior occurs because in this case nucleation on top of existing islands is strongly enhanced by the Schwoebel barrier. In fact, as Fig. 4 indicates with increasing Schwoebel barrier coefficient v_1 (in absolute magnitude) the pinning induced oscillations are further amplified. Note, however, that calculations of $\Delta T_c/T_c$ in terms of Eq. (2) are limited to sufficiently small coefficients v_1 in order that the rms roughness amplitude $w(\propto [\int\langle|h(q,t)|^2\rangle \mathrm{d}^2q]^{1/2})$ to remain significantly smaller than the film thickness d (Fig. 5)

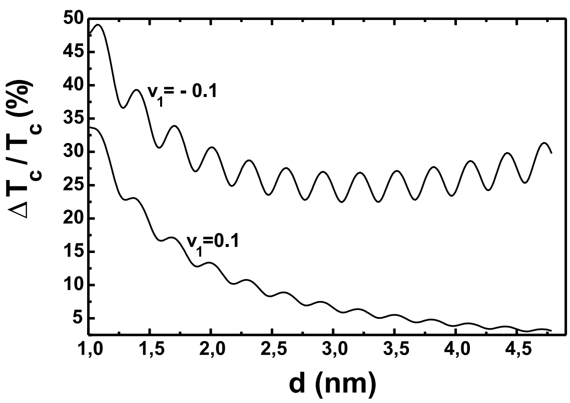

Fig. 3. $|\Delta T_c|/T_c$ vs. film thickness d where comparison of stable and unstable Q-LBL is shown for pinning amplitude $A = 0.3$ and relaxation coefficients $v_1 = \pm 0.1$.

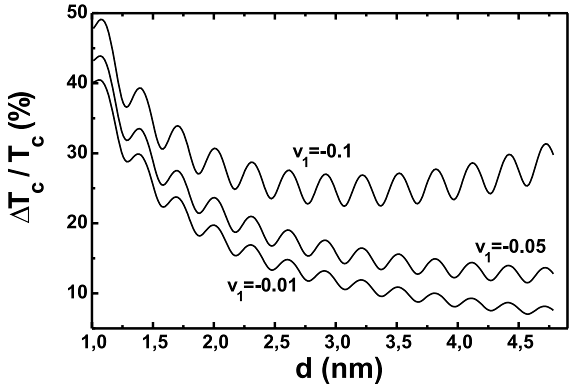

Fig. 4. $|\Delta T_c|/T_c$ vs. film thickness d for unstable Q-LBL growth for pinning amplitude $A = 0.3$ and relaxation coefficients v_1 as indicated.

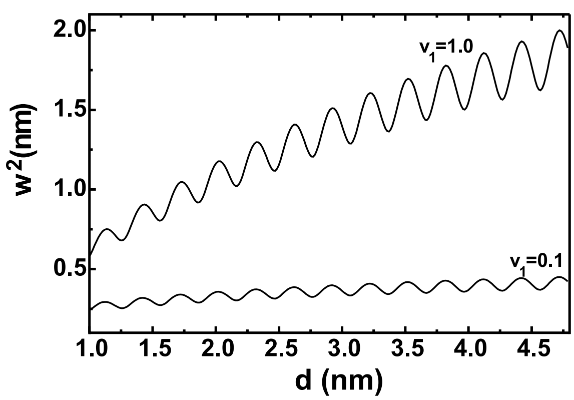

Fig. 5. Squared roughness amplitude w vs. d for stable growth with $A = 0.3$, $D = 0.02$ nm s^{-1}, and v_1 as indicated.

[15,16]. In general, the rms roughness amplitude will grow with increasing film thickness in an oscillatory manner with amplitude that depends on the pinning amplitude A (Fig. 5).

Finally, we should point out that different growth techniques (i.e., thermal/e-beam evaporation, molecular-beam epitaxy, pulsed laser deposition, sputtering, etc.) will result in different growth scenarios (self-affine fractal roughness, mound roughness, Q-LBL etc.) [8–10,18]. Therefore, in general, the thickness dependence of the local surface slope will be different for different growth modes, which will result in different morphology contributions on proximity pinning effects [7,16].

4. Conclusions

We investigate the influence of Q-LBL interface roughness on proximity effects, at the junction of a thin surperconducting/normal-metal film. The SN junction is assumed to be formed on a superconducting film for which the film surface was grown at a first place in a Q-LBL mode. The reduction of the superconducting film critical temperature $\Delta T_c/T_c$ is shown to decrease with film thickness in an oscillatory manner which reflects the presence of growth front pinning effects during roughness formation. The oscillations are significant for weak surface relaxation where $\Delta T_c/T_c$ varies significantly with film thickness, and decrease in magnitude with increasing film thickness. In contrast, in the presence of growth front instability, the pinning induced oscillations are shown to be further amplified with increasing instability strength.

Although up to now precise experimental investigation of roughness influence on proximity effects is rather limited [5], recent studies of systems (i.e., Pb films [18–20]) grown closely in a Q-LBL mode have indicated the possibility of similar investigations on proximity effects. Nevertheless, besides growth front aspects, further studies will be required to incorporate on proximity effects changes of the interface morphology because of possible stress that might develop during the SN junction formation.

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Appendix A

Outline for derivation of Eq. (3): if we assume the perturbative expansion $h = h_1 + \varepsilon h_2 + \varepsilon^2 h_3 + \cdots$, Eq. (2) yields, i.e., for $n = 1$.

$$\begin{aligned} \frac{\partial h_1}{\partial t} &= R + v\nabla^2 h_1 + n, \\ \frac{\partial h_2}{\partial t} &= v\nabla^2 h_2 - A\sin\left(\frac{2\pi h_1}{c}\right) + n. \end{aligned} \tag{A.1}$$

Setting $h_1 = h_{1,0} + h_{1,1}$ with $h_{1,0} = Rt$, Eq. (A.1) yields $h_{1,1}(\mathbf{q}, t) = \int_0^t n(\mathbf{q}, \tau)\mathrm{e}^{-(kq^4+vq^2)(t-\tau)}\,\mathrm{d}\tau$ with $h_{1,0}$ the average surface height and $h_{1,1}$ the height fluctuation at zero order perturbation. Since $\langle h_{1,1}^2\rangle^{1/2} \ll h_{1,0}$, the pinning sine-term in Eq. (A.1) can be further approximated by

$$\frac{\partial h_2}{\partial t} = v\nabla^2 h_2 - A\sin(t/\tau_c) - \frac{2\pi A}{c} h_{1,1}\cos(t/\tau_c) \tag{A.2}$$

with $\tau_c = c/2\pi R$. If we set $h_2 = h_{2,0} + h_{2,1}$ such that

$$\begin{aligned} \frac{\partial h_{2,0}}{\partial t} &= -A\sin(t/\tau_c), \\ \frac{\partial h_{2,1}}{\partial t} &= v\nabla^2 h_{2,1} - \frac{2\pi A}{c} h_{1,1}\cos(t/\tau_c) \end{aligned} \tag{A.3}$$

and integrate, we obtain the average film thickness $\langle h\rangle = h_{1,0} + h_{2,0}$ or $\langle h\rangle = Rt + A\tau_c[\cos(t/\tau_c) - 1]$ and

$$h_{2,1}(\mathbf{q}, t) = -A\int_0^t h_{1,1}(\mathbf{q}, \tau)\cos(\tau/\tau_c)\mathrm{e}^{vq^2(t-\tau)}\,\mathrm{d}\tau, \tag{A.4}$$

which finally yields the roughness spectrum of Eq. (3).

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